Effect of number of layers on the compression behaviour of graphenes

Ch. Androulidakis^{1,2}, *E.N. Koukaras*¹, *G. Tsoukleri*¹, *D. Sfyris*¹, *I. Polyzos*¹, *J. Parthenios*¹, *K. Papagelis*¹, and *C. Galiotis*^{1,3,*}

¹ 1 Institute of Chemical Engineering and High Temperature Chemical Processes,

Foundation for Research and Technology - Hellas (FORTH), P.O. BOX 1414, Patras 265 04, Greece

² Department of Materials Science, University of Patras, Patras 26504, Greece

^{2*}Affiliation of the corresponding author: FORTH/ ICEHT and Univ. of Patras, PO BOX 1414, Patras 26504, Greece

Graphene has received a lot of attention nowadays due to the fact that as a single, virtually defectfree crystal is predicted to have an intrinsic tensile strength higher than any other known material¹. Regarding tension, the original experiments employed bending of suspended flakes on an AFM apparatus have yielded, with the help of adequate modelling and a number of assumptions, values of tensile strength of 130 GPa. So far however, these values have not been confirmed by direct uniaxial or biaxial experiments. In compression, however, it is clear that thin monoatomic membranes have no practical resistance to compression. This picture however changes dramatically when graphenes are embedded in polymer matrices such as PMMA. As shown in our recent publications^{2,3} exfoliated monolayer graphene exhibits a compression strain to first failure (Euler buckling) of 0.6% which is indeed a very high value for its overall dimensions (typically 30 by 20 μ m). Furthermore, the obtained values are not affected by the flake size and can be modelled quite adequately by considering Winkler-type of mechanics upon loading^{2,3}.

In this work, graphene flakes with thickness of 1 to 3 layers were tested under axial compression on a four-point-bend rig. In figure 1a-c selected results from the compression experiments are presented. By monitoring the position of the 2D Raman peak versus applied strain we can estimate the point of elastic (buckling) or yield (cohesive) failure of the various specimens. Moreover, the slope of the Raman frequency vs. strain is indicative of the stress transfer efficiency of the system. Initially as expected phonon hardening at a slope of $-56 \text{ cm}^{-1/\%}$ is observed. As the compressive strain increases, a plateau is reached which corresponds to the failure of the graphene while further loading causes phonon softening. As mentioned above, the monolayer graphenes failed at a critical strain of -0.6% with Euler-type of elastic buckling of failure¹. However, as the number of layers increases the critical strain to failure was found to decrease. Since the thickness of the graphene specimens increases with number of layers one would expect for an elastic instability the inflection point is shifted to higher strains. The fact that a notable decrease of the plateau strain (inflection point) is observed indicates that the mode of failure is different than that observed for a monolayer. In particular, the bilayer graphene failed at a strain of -0.25% and the trilayer at a strain of -0.20%. This behaviour can be attributed to the weak interlayer bonding between the individual graphene layers. In fact the bonding between the graphene outer surfaces and the polymer matrix is estimated to be higher than that of the internal graphene-graphene bonding and cohesive failure is promoted prior to Euler buckling. We conclude therefore that in multilayer graphene a lower threshold of critical buckling strain is obtained due to the cohesive failure within the material. The results of the present study provide crucial insight for the effective design of graphene nanocomposites under compression and show that monolayer graphene offers higher resistance to compressive failure than two or three layered graphene materials.

References

- [1] C. Lee et al., *Science* 321, p. 385, (2008).
- [2] Frank et al, ACS Nano, 3131–3138: 4/6, 2010
- [3] Ch. Androulidakis et al., *SRep* (*Nature*). 4, (2014).

³ Department of Chemical Engineering, University of Patras, Greece

Acknowledgement

This research was supported by This research has been co-financed by the European Union (European Social Fund-ESF) and Greek national funds through the Operational Program "Education and Life long Learning" of the National Strategic Reference Framework (NSRF) - Research Funding Program: ERC-10 "Deformation, Yield and Failure of Graphene and Graphene-based Nanocomposites". Finally, the authors acknowledge the financial support of the Graphene FET Flagship ("Graphene-Based Revolutions in ICT And Beyond"- Grant agreement no: 604391) and of the European Research Council (ERC Advanced Grant 2013) via project no. 321124, "Tailor Graphene".



Figure 1: Position of the 2D Raman peak versus the applied strain for (a) monolayer, (b) bilayer and (c) trilayer graphene respectively.